Effects of neutron irradiation on glass ceramics as pressure-less joining materials for SiC based components for nuclear applications

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Abstract

This paper reports on the microstructure and properties of two glass-ceramics based on SiO_2-Al_2O_3- MgO (SAMg) and SiO_2-Al_2O_3-Y_2O_3 (SAY), which have been designed to be used as pressure-less low activation joining materials for SiC/SiC and SiC based components for nuclear applications. Glass-ceramic pellets (SAY and SAMg) were irradiated for approximately one year in the reactor core of the LVR-15 research reactor at Nuclear Research Institute Rez, Czech Republic, at about 50 °C , $6.92 \cdot 10^{24} n/m^2$ (E>1 MeV, about 1 dpa in steel); SiC/SiC composites joined by SAY were irradiated about one year at HFR (High Flux Reactor) , Petten, The Netherlands, 550°C, $9\text{-}11 \cdot 10^{24}$ n/m^2 (E>1 MeV, about 1.4- 1.8 dpa in C), 600 °C, $16\text{-}22 \cdot 10^{24}$ n/m^2 (E>1 MeV, about 2.6-3.3 dpa in C) and 820 °C $31\text{-}32 \cdot 10^{24}$ n/m^2(E>1 MeV, about 5 dpa in C). Optical microscopy with image analysis and scanning electron microscopy (SEM) with X-ray microanalysis (EDS) were used to investigate the glass-ceramics morphology and composition, showing a remarkable similarity before and after neutron irradiation for both glass-ceramics. Comparison of bending strength for irradiated and non-irradiated SAY joined SiC/SiC indicate that the mechanical strength is unaffected by irradiation at these conditions.

Keywords

Joining, SiC, SiC/SiC, glass-ceramic, neutron irradiation

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Introduction

Ceramic fibre reinforced ceramic matrix composites (CMC) are promising materials for nuclear applications and require extreme temperature stability and neutron irradiation resistance. Composites based on SiC fiber reinforced SiC matrix (SiC/SiC) have significantly higher resistance to high temperatures and aggressive environment in comparison to metals or other materials. Another advantage for nuclear applications is their high resistance to thermal shock and low induced radioactivity [1].

One long-standing practical issue with regard to the application of SiC/SiC for nuclear systems is the joining of SiC/SiC to obtain complex components, because they cannot be connected together by ordinary welding (there is no melting phase) or by self diffusion bonding (diffusion is very slow even at high temperature). In the field of nuclear energy production, requirements for SiC/SiC joints and joining methods are extremely severe: advances on this topic is considered a useful technology development for SiC-based CMC. Joining materials must fulfill several requirements: coefficient of thermal expansion must be close to that of SiC/SiC, it must have good wettability to SiC/SiC, pressure-less and localized heating joining techniques are preferred, it must have shear strength satisfying the design requirement, it must have acceptable transmutation reactions and low neutron-induced radioactivity, it must be chemically compatible with coolant (He or LiPb), fuel, and fission products and other functional materials [2].

Only a limited number of materials for bonding of SiC/SiC composites to be used in nuclear reactors have been proposed [3, 4, 5], though none of them have been tested in nuclear environment. One such material, utilizing the NITE technology (nano-infiltration transient eutectic), where the transient eutectic is a liquid phase based on Y_2O_3 and Al_2O_3, (initially also SiO_2), has been successfully used to join SiC based components: to the best of the authors' knowledge, no results have been published on neutron irradiated NITE joints, but TEM analysis of NITE SiC/SiC composite using dual-ion irradiation gave promising results [6]. A few glass-ceramics with compositions tailored to achieve the above requested properties are currently under evaluation as

pressure-less joining materials for nuclear environment [7-10]; SiO_2-Al_2O_3-Y_2O_3(SAY) and SiO_2-Al_2O_3- MgO (SAMg) glass-ceramics, discussed in [7, 11], have been proposed as potential low activation, pressure-less joining materials for SiC based components for nuclear applications, but never tested in a nuclear environment up to now.

The use of glass-ceramics as joining materials in a neutron environment is still a matter of discussion and scepticism within the scientific community. While several papers report on the interaction of X- or gamma-rays with glass and glass-ceramics, a few references are available on neutron irradiation of glass ceramics.

Some references [12, 13] reported the swelling (about 15 vol%) of crystalline silica at fluence of $2 \cdot 10^{24}$ n/m^2 and the densification of vitreous silica (about 3 vol%). In [14] the changes in the optical, physical and electrical properties of quaternary silicate glasses under neutron fluence from $1 \cdot 10^{18}$ n/m^2 to $1 \cdot 10^{21}$ n/m^2 and the observed changes in glass network structure are discussed. Neutron irradiation effects on glass have been discussed in terms of glass shrinkage, mainly due to the formation of thermal spikes, and glass dilation due to atom displacement and bond cracking in [15, 16, 17] the effect of thermal cycling and neutron radiation up to 10^{22} n /m^2 on the microstructure of a fused silica-stainless steel interface of a viewport for ITER is investigated, but no results are presented on glass microstructure after neutron irradiation.

Reference [18] studied a glass-ceramic (MACOR, Corning) made of 50 % vol. fluorophlogopite mica crystals ($KMg_3AlSi_30_{10}F_2$) embedded in a borosilicate glass matrix, at room temperature, with fluences ranging between $1 \cdot 10^{20}$ and $1 \cdot 10^{22}$ n/m^2. In Ref. [19] D.L. Porter et al. characterized eight glass-ceramics containing $KMg_3AlSi_30_{10}F_2$ or $Li_2Si_2O_5$ in a glass matrix, at 400-500 °C, with a fluence of $2.4 \cdot 10^{22}$ n/m^2. Ref. [20] observed expansion of the mica phase and contraction of the glass matrix in a MACOR glass-ceramic at room temperature, with a fluence of $1 \cdot 10^{23}$ n/m^2.

Two competing processes such as swelling and densification of amorphous and crystalline phases present in a glass-ceramic have been observed in the above mentioned glass-ceramics, with a

differential swelling/shrinking of two phases which typically lead to a reduction of strength, separation of phases, and failure of the glass-ceramic.

Experimental part

SAY and SAMg glass ceramics

SiO_2-Al_2O_3-Y_2O_3(SAY) and SiO_2- Al_2O_3- MgO (SAMg) were designed as low activation materials, as simulated by European Activation System EASY-2007 code package [21] and discussed in [7]; they were prepared by melt quenching by oxides and carbonates starting products as described in [7, 11, 22].

SAY and SAMg parent glasses were ground and sieved at size 38<μm then sintered in a tubular oven in Ar flow (heating rate of 1000 °C/h during the heating, 6° C/h during cooling) at 1375 °C for 20 min then 1235 °C for 1 hour to obtain SAY glass-ceramic and at 1180 °C, 1 hour for SAMg glass-ceramic.

The sintering process was the same one used to join (or coat) SiC/SiC by these glass-ceramics, as discussed in the next paragraph.

The coefficient of thermal expansion (CTE) measured between 400 and 700°C is $5.5 \cdot 10^{-6}$ $°C^{-1}$for SAY glass ceramic [7] and $3 \cdot 10^{-6}$ $°C^{-1}$ for SAMg glass-ceramic measured between 200 and 600 °C [11], both close to that of SiC/SiC, $4 \cdot 10^{-6}$ $°C^{-1}$ at room temperature [7].

Both glass-ceramics showed very good wettability to the SiC/SiC surface, measured by hot stage microscopy (Leitz GmbH AII) equipped with a Leica DBP (Ernst Leitz GMBH, Wetzlar, Germany) camera: SAY contact angle was close to zero at 1400 °C [7] whereas a temperature of 1480 °C was necessary to reach zero contact angle for SAMg [11].

Crystalline phases and characteristic temperatures of both glass-ceramics have been measured and discussed in [7].

Two pellets (diameter=8mm, height= 4 mm) of sintered SAMg and SAY glass ceramics were characterized by optical microscopy and SEM-EDS (Nikon Epiphot 300 with digital camera DVC and Tescan Vega TS 5130 XM with X-ray microanalysis module EDS).

Evaluation of sample porosity was carried out using light microscope micrographs; the image analysis was carried out using program NIS – Elements AR 3.10. Microhardness measurement was done according to Vickers – MHV 0.1 using Anton Paar MHT-4 instrument.

Neutron irradiation conditions for SAMg and SAY glass-ceramic pellets were about 50 °C measured by putting a thermocouple directly in the reactor water, $6.92 \cdot 10^{24} n/m^2$ (E>1 MeV, about 1 dpa in steel).

The joined samples were irradiated for about one year at HFR, Petten, NL, at 550°C, $9\text{-}11 \cdot 10^{24}$ n/m^2 (E>1 MeV, about 1.4-1.8 dpa in C), at 600 °C, $16\text{-}22 \cdot 10^{24}$ n/m^2 (E>1 MeV, about 2.6-3.3 dpa in C) and 820 °C $31\text{-}32 \cdot 10^{24}$ n/m^2 (E>1 MeV, about 5 dpa in C).

The nominal temperatures were 550°C, 600°C and 900°C. Because the experiment was aimed at screening new materials rather than providing design data of those materials, a relative large tolerance (± 30 °C) was considered acceptable.

The samples were placed in sample holders (drums), surrounded by an inert gas-mixture (helium and/or neon). The atmosphere inside the sample holder is a gas mixture of helium and/or neon in stagnant conditions; the experiment capsule with the drums inside was purged with gas mixture and then sealed. No oxygen nor humidity was detected. All drums are made of a molybdenum alloy (TZM, 0.5Ti, 0.08Zr, Mo). The high density of TZM allowed to reach the required sample temperatures of 550°C, 600°C and 900°C. The temperature control was possible due to a combination of nuclear heating, different gas gap width in the sample holder and a certain gas mixture (helium and neon).

The instrumentation of the sample holders consisted of 23 thermocouples. All thermocouples were of type N, with Inconel 600 sheathes of 1 mm outer diameter and MgO as insulator. The thermocouples were used to continuously monitor and record the temperature of the specimens in

the sample holder. Tuning the temperature of the experiment during neutron irradiation was done by changing the gas mixture in the second containment until the required temperature was reached. Irradiated samples were handled in semi-hot metallographic cell: samples were cut by a diamond saw in two parts then ground and polished (OPS Struers). For SEM observation the samples were sputtered by carbon (AGAR TURBO CARBON COATER).

SAY joined SiC/SiC

SiC/SiC has been provided by MT Aerospace AG, (Germany): it is a 2D SiC/SiC composite, with Tyranno Fiber S SiC fibers (UBE industries), crystalline Chemical Vapour Infiltration (CVI) SiC matrix and a few µm thick pyrolitic carbon interface [7]. The composition of the SiC fiber (% wt) is the following: Si= 50.4; C=29.7; O=17.9 ;Ti=2.0; the diameter is 8.5-11 µm. The SiC/SiC composite has a 0°/90° plain weave fabric, which is parallel to the mechanical loads used to characterize the joined samples.

SAY has been used to join SiC/SiC as described in [7] with a pressure-less slurry based joining technique (Ar flow at 1375 °C for 20 min then 1235 °C for 1 h). Non-flat SiC/SiC were joined by SAY as described in [7] and briefly sketched in Table 1: Type 2 is a modification of the Mortise and Tenon joint configuration [23] and Type 3 geometry is a half-lap joint [24,25, 26].

SAY joined SiC/SiC were tested by four point bending before [7] and after neutron irradiation. Four-point bending strength was calculated using 2.6 mm x 5.2 mm x 45 mm samples (sample size adapted from ASTM C1341-00; support span: 40 mm; load span: 20mm.

Irradiation was performed within the EU-Project Extremat (http://www.extremat.org/) at the High Flux Reactor (HFR) Petten. Irradiated samples have been investigated in the NRG Hot Cell Laboratory (HCL) in Petten, the Netherlands.

Neutron irradiation conditions and bending strength results on SiC/SiC and SAY joined SiC/SiC are shown in Table 1: standard deviation is provided for results obtained on at least five samples. When absent, it means that only one sample has been measured. The asterisk means irregular failure, not in compliance with the standard ASTM C1341.

The glass ceramic joined SiC/SiC was observed before and after neutron irradiation by optical microscopy using a Leitz MM 5 RT Light Microscope and SEM-EDS with a JEOL 6490 LV SEM.

Results and discussion

SAY and SAMg glass ceramics

SAY

The SAY glass-ceramic resulted in a porous microstructure after sintering (pellets) (Figure 1a) and after pressure-less joining by slurry (Figure 5 a): this was due to a non-optimized thermal treatment. Recent improvements of the process have yielded a much lower porosity, though there are still no neutron irradiation results on this newer, denser microstructure.

As shown on Figure 1 b, SAY darkened in colour after irradiation, due to a change of optical properties of the material, such as irradiation induced defects typical of glasses and glass-ceramics [14, 26] .

The average chemical composition of SAY before and after irradiation measured by EDS is given in Table 2 . SAY composition remains unchanged after irradiation, corresponding to the as cast one and fulfills the condition of low-induced activity (Al < 18 wt %) [7].

As expected from the silica–alumina–yttria phase diagram and as already reported for non-irradiated SAY glass-ceramic in [22, 27] three phases were identified by TEM - SAED as SiO_2 (cristobalite),diyttriumdisilicatekeiviite$Y_2Si_2O_7$ and aluminium silicon oxide (non-stoichiometric mullite). Additionally, very small Y_2O_3 particles (30 nm) were present in the $Y_2Si_2O_7$ grains. In this work (fig 2), SEM-EDS compositional analysis on non-irradiated (Figure 2a) and irradiated (Figure 2b) SAY glass-ceramic pellets confirms the morphology and composition of three phases: a black phase (cristobalite, SiO_2), a white needle-like phase ($Y_2Si_2O_7$, keiviite) and a grey phase (mullite, Al_2SiO_5), with a remarkably similar morphology of both samples. An approximate area ratio of the three phases has been measured by image analysis and found unchanged before and

after irradiation: grey phase (64.7 wt %, Al_2SiO_5), black phase (35 wt %, SiO_2) and white needle-like phase (5.3 wt %, $Y_2Si_2O_7$). No evidence of dimensional or density change have been measured after irradiation on these samples.

SAMg

The average chemical composition of SAMg before and after irradiation measured by EDS is given in Table 3 : it corresponds to the as cast glass (and fulfills the condition of low-induced activity (Al < 18 wt %) [11].

As shown on Figures 3a and 3b, also SAMg darkened after irradiation. SEM-EDS compositional analysis on non-irradiated and irradiated SAMg pellets confirms the same element composition for both samples. The poor contrast of phases (Figure 4) did not allow the measurement of phase ratio by image analysis, but as reported in [11] the SAMg glass-ceramic has a 3:1 cordierite to mullite structure with negligible residual amorphous phase.

Porosity and micro-hardness have been measured on irradiated and non-irradiated SAY and SAMg samples: values are not reported here since the porosity of these samples is too high to give relevant information for these glass-ceramics, due to reasons discussed above on the not optimized sintering process. No evidence of dimensional or density change have been measured after irradiation on these samples.

Finally, it must be noted that micro-cracking due to differential swelling/shrinkage of different phases as reported in [20] for MACOR glass-ceramics have not been observed in SAY and SAMg glass-ceramic at these irradiation conditions. A similar behavior of Y_2O_3-Al_2O_3 and MgO-Al_2O_3 based ceramics have also been reported by [28] on polycrystalline pellets of yttrium aluminate garnet ($Y_3Al_5O_{12}$) and magnesium aluminate spinel ($MgAl_2O_4$) after irradiation in the high flux reactor (HFR) at Petten, NL, to a neutron fluence of $1.7 \cdot 10^{26}$ n/m^2 (E > 0.1 MeV) at a temperature of about 542 °C. Volume changes smaller than 1% have been measured for $Y_3Al_5O_{12}$ and $MgAl_2O_4$.

SAY joined SiC/SiC

The SiC/SiC coupons were pressure-less joined by SAY by a slurry based joining process in Ar flow at 1375 °C for 20 min then 1235 °C for 1 h: room temperature bending tests on non-irradiated samples have been reported in [7] for Type 2 and Type 3 and are summarized in Table 1. For comparison purposes, Table 1 shows also the bending strength of the as received non joined SiC/SiC and of the non joined SiC/SiC heated at 1420 °C, 30 minutes, then at 1240 °C, one hour, in Ar flow, at a slightly different conditions from those used for the joining process: as it can be seen, a severe decrease of bending strength was observed on the non joined SiC/SiC after this heat treatment, due to the unsuitability of Tyranno Fiber S SiC fibers to withstand these temperatures. Unfortunately, it was impossible for the authors to obtain higher temperature resistant SiC/SiC and the activity was continued on these composites. The joining process further reduced the SiC/SiC bending strength for both Type 2 and 3, but Type 2 mostly showed the failure in the composite instead of in the joined area, thus suggesting a possible higher bending strength in case of more resistant SiC/SiC and a less porous joining material.

As received not joined SiC/SiC also showed a decrease in mechanical properties after irradiation (Table 1): its bending strength was severely reduced after irradiation at 550 °C, 9-11·10^{24} n/m^2 and even lower values were measured at 600 °C, 16-22·10^{24} n/m^2 and at 820 °C, 31-32·10^{24} n/m^2. The asterisk in Table 1 for these latter results indicates that an irregular bending failure not in compliance with the standard ASTM C1341 was obtained, e.g. some samples failed in a shear mode rather than a tensile mode.

However, all SAY joined SiC/SiC were still joined after irradiation and were submitted to bending tests. All SAY joined SiC/SiC samples failed in accordance with the standard ASTM C1341, and Table 1 shows their measured bending strength. A statistical analysis had been possible only for SAY joined SiC/SiC type 2 at higher irradiation temperature, 820 °C and fluence, 31-32·10^{24} n/m^2, where 89±17 MPa have been measured on four samples. All other cases where an average value is

not available represent measurements obtained up to now on one sample and should be considered as a first indicative behavior.

An encouraging 118 MPa has been obtained with fracture in the joining material for type 2 SAY joined SiC/SiC (600 °C 16-22·10 ^{24} n/m^2), comparable to 122 ± 10 MPa measured on the non-irradiated joined sample. Much lower results (89±17 MPa) have been measured for type 2 at higher irradiation temperature, 820 °C and fluence, 31-32·10 ^{24} n/m^2. These are not directly comparable to values measured on non-joined SiC/SiC at the same irradiation conditions because they failed with an irregular failure, not in compliance with the standard ASTM C1341, as in Table 1.

It must be underlined that one of the design for a DEMO fusion reactor, TAURO [25 Figure 1.TAURO blanket design, detail A] has a type 3 geometry for the SiC/SiC structure of the reactor: thus, type 3 SAY joined SiC/SiC after irradiation can be considered as the closest SiC/SiC based mock-up component ever built of a DEMO reactor.

Bending strength of one sample type 3 SAY joined SiC/SiC after irradiation at 600 °C, 16-22·10 ^{24} n/m^2 , gave 65 MPa, which is much lower than for the non-irradiated SAY joined SiC/SiC, but not statistically relevant. However, the same glass-ceramic in the same irradiation condition gave an encouraging 118 MPa for type 2 SAY joined SiC/SiC, comparable to 122 ± 10 MPa measured on the non-irradiated joined sample: the role of random porosity in the joining material might be the reason for these results. Further tests must be done with a reduced porosity in the joined area.

From a morphological and compositional point of view, SAY joined SiC/SiC showed again an encouraging behavior: Figure 5 shows SEM of SAY joined SiC/SiC (type 3) before (a) and after (b) irradiation (HFR-Petten) at 600 °C, 16.3·10 ^{24} n/m^2 and bending tests: as discussed before, for SAY pellets irradiated at LVR-15 (Figure 1 and 2), a random and excessive porosity was present in the joining material, which was in any case able to withstand bending test.

A closer look (Figure 6) clearly shows that the microstructure as observed by SEM of SAY joined SiC/SiC before (Figure 6 a) and after (Figure 6 b) irradiation (HFR-Petten) at 600 °C, 16.3·10 ^{24} n/m^2 did not yield observable microstructural changes. The same morphology has been found for

type 2 and type 3 samples in their SAY joined areas. Three phases can be observed in Figure 6 (higher magnification) before and after irradiation; their composition, measured by EDS (not reported here), corresponds to the three phases discussed above. Morphology of SAY joined SiC/SiC after irradiation at 820 °C, 31-32·10^{24} n/m^2 are not available yet.

Porosity has been observed on non-irradiated (Figure 6 a) and irradiated (Figure 6b) samples without yielding any observable changes.

Again, it must be underlined that differential swelling/shrinkage of different phases reported in [20] for MACOR glass-ceramics have not been observed on SAY glass-ceramic joints at these irradiation conditions, as summarized in Table 4.

Conclusions

The aim of the work was to evaluate microstructure and compositional behavior of irradiated glass-ceramics used to join SiC/SiC and compare these results with values for non-irradiated samples.

Both glass-ceramic joining materials (SAY, SAMg) darkened after irradiation because irradiation probably changed the optical properties of glass-ceramics. Within the statistical limitations of this study, this was the only observable effect of irradiation on the joints observed. Results indicate that irradiation at 50 °C, 6.92·10^{24} n/m^2 does not affect the chemical composition of the glass-ceramic materials.

In SAY glass-ceramic the same three phases are present before and after irradiation: mullite, SiO_2 and needle-like $Y_2Si_2O_7$. The same glass-ceramic microstructure (i.e. fracture surfaces, macro and micro-pores, etc.) has been observed before and after irradiation for SAMg glass-ceramic.

Likewise, no significant irradiation effects on porosity were observed.

The differential swelling/shrinkage of different phases previously reported in [20] for MACOR glass-ceramics have not been observed on SAY and SAMg glass-ceramic at these irradiation conditions, which would have been potentially devastating to the applicability of this joint material for nuclear application.

The microstructure observed by SEM of SAY joined SiC/SiC before (a) and after (b) irradiation (HFR-Petten) at 600 °C, $16.3 \cdot 10^{24}$ n/m^2 likewise did not change in a detectable way.

The SAY joined SiC/SiC materials of this study remained intact and were apparently unaffected by irradiation at 600 °C, $16.3 \cdot 10^{24}$ n/m^2 and at 820 °C, $31\text{-}32 \cdot 10^{24}$ n/m^2. When submitted to bend testing, type 2 SAY joined SiC/SiC resulted in an encouraging 118 MPa comparable to 122 ± 10 MPa measured on the non-irradiated joined sample.

In conclusion, if previous works have not been encouraging at low dose, these results can be considered promising for reactor designs such as TAURO, or in general when a transition from SiC based components to SiC or to other materials is necessary, away from the first wall. Further improvements are expected by testing the improved low porosity glass-ceramic joining materials to their lifetime dose.

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References

[1] L.L. Snead, T. Nozawa , M. Ferraris , Y. Katoh , R. Shinavski , M. Sawan, Silicon carbide composites as fusion power reactor structural materials, J. Nucl. Mater. (2011), in press doi:10.1016/j.jnucmat.2011.03.005.

[2] M. Ferraris, M. Salvo, V. Casalegno, S. Rizzo, A. Ventrella, Joining and integration ssues of ceramic matrix composites for nuclear applications in Processing and Properties of Advanced Ceramics and Composites II , Ceramic Transactions (2) Edited by Narottam P. Bansal, Jitendra P. Singh, Jacques Lamon Sung, R. Choi, Morsi M. Mahmoud (2010).

[3] T. Hinoki, N. Eiza, S. Son, K. Shimoda, J. Lee, A. Kohyama, Development of joining and coating technique for SiC and SiC/SiC Composites utilizing NITE processing, Ceram. Eng. Sci. Proc. 26 (2005) 399-405.

[4] H-C. Jung, Y-H. Park, J-S. Park, T. Hinoki, A. Kohyama, R&D of joining technology for SiC components with channel, J.Nucl. Mater. 386-388 (2009) 847-851.

[5] C.H. HenagerJr, Y. Shin, Y. Blum, L.A. Giannuzzi, B.W. Kempshall, and S.M. Schwarz, Coatings and joining for SiC and SiC-composites for nuclear energy systems , J. Nucl.Mater. 367-370 (2007) 1139-1143.

[6] H. Kishimoto K. Ozawa , O. Hashitomi, A. Kohyama, Microstructural evolution analysis of NITE SiC/SiC composite using TEM examination and dual-ion irradiation, J. Nucl. Mater 367–370 (2007) 748–752.

[7] M. Ferraris, M. Salvo, .V. Casalegno, A. Ciampichetti, F. Smeacetto, M. Zucchetti, Joining of machined SiC/SiC composites for thermonuclear fusion reactors. J. Nucl. Mater. 375 (2008) 410-415.

[8] M. Ferraris, M. Salvo, V. Casalegno, S. Han, Y. Katoh, H.C. Jung, T. Hinoki, A. Kohyama, Joining of SiC-based materials for nuclear energy applications, J. Nucl. Mat., in press doi:10.1016/j.jnucmat.2010.12.160 (2011) .

[9] Y. Katoh, M. Kotani, A. Kohyama, M. Montorsi, M. Salvo, M. Ferraris, Microstructure and mechanical properties of low activation glass-ceramic joining and coating for SiC/SiC composites. J. Nucl. Mater. 283-287 (2000) 1262-1266.

[10] Mailliart, V. Chaumat, F. Hodaj, French Patent n. 08 55857 filed on Sept 2008.

[11] M. Ferraris, M. Salvo, F. Smeacetto , Cordierite–mullite coating for SiC_f/SiC composites, J. Eur. Ceram. Soc. 22 (2002) 2343–2347.

[12] M. Wittels, F. A.Sherrill, Radiation damage in $Si0_2$ structures, Phys. Rev. 93 (1954) 1117-1118.

[13] W. Primak , L. H. Fuchs, and P. Day, Effects of nuclear reactor exposure on some properties of vitreous silica and quartz, J. Am. Ceram Soc. 38 (1955) 135-9.

[14] A.K. Sandhu, S. Singh, O.P. Pandey, Neutron irradiation effects on optical and structural properties of silicate glasses , Mater. Chem. Phys. 115 (2-3) (2009) 783.

[15] M.M. Morsi, S. El-Konsol, M.A. Adawi, Effect of neutron and gamma irradiation on some properties of borate glasses, J. Non- Cryst. Solids , 58 (2-3), (1983) 187 .

[16] D. Kenji , Structure changes in amorphous silica by neutron irradiation, J. Non-Cryst. Solids, 51 (3), (1982) 367.

[17] M. Jacobs, G. Van Oost, J. Degrieck, A. Gusarov, V.Massaut, M. Schyns, Microstructural effects of temperature and neutron irradiation on optical windows, J. Nucl.Mater, in press (2011), doi:10.1016/j.jnucmat.2010.12.145.

[18] J. D. Fowler, Jr., C. F. Hurley, J. C. Kennedy, and F. W. Clinard, Jr., 14 MeV Neutron irradiation effects in MACOR glass-ceramics, J. Nucl.Mater, 103-104, (1981) 755.

[19] D. L. Porter, M. R. Pascucci, B. H. Olbert, Neutron irradiation effects on SiO_2 and SiO_2 -based glass ceramics, J. Nucl.Mater, 103- 104 (1981) 767-772.

[20] W.A. Coghlan, F.W. Clinard, Damage to Macor glass-ceramic from high-dose 14 MeV neutrons, J. Nucl.Mater , 179-181 (1991) 391-394.

[21] R.A. Forrest, The European Activation System: EASY-2007 Overview, UKAEA Report UKAEA FUS 533, January 2007.

[22] F. Smeacetto , M. Ferraris , M. Salvo , S.D. Ellacott , A. Ahmed , R.D. Rawlings , A.R. Boccaccini, Protective coatings for carbon bonded carbon fibre composites, Ceram. Int., 34 (2008) 1297 – 1301.

[23] H. Tokgoz, A. Ozcifci, M. Atar, B. Uysal, Shear and bending strength of some end to end grained joints prepared from scotch pine, Turkish Journal of Agriculture and Forestry 23 (1999) 621-625.

[24] R.L. Bruce, S.K. Guharay, F. Mako, W. Sherwood, E. Lara-Curzio. Polymer derived SiC_f/SiC_m composite fabrication and microwave joining for fusion energy applications. Proceedings of the 19th IEEE/IPSS Symposium on Fusion Engineering, Atlantic City (NJ) January 21-25 (2002) 426-429.

[25] H. Golfier, G. Aiello, M. Futterer, L. Giancarli, A. Li-Puma, Y. Poitevin a, J. Szczepanski, Progress on the TAURO blanket system Fusion Engineering and Design 61-62 (2002) 461-470.

[26] N.A. El-Alaily , R.M. Mohamed, Effect of irradiation on some optical properties and density of lithium borate glass, Mater. Sci. Eng. B98 (2003) 193-203.

[27] F. Smeacetto, M. Salvo , M. Ferraris , V. Casalegno , G. Canavese, T. Moskalewicz, S. Ellacott, R. D.Rawlings , A. R. Boccaccini, Erosion protective coatings for low density, highly porous carbon/carbon composites, Carbon, 47 (2009) 1511-1519.

[28] E.A.C. Neeft, R.J.M. Konings, K. Bakker, J.G. Boshoven, H. Hein, R.P.C. Schram , A. van Veen, R. Conrad, Neutron irradiation of polycrystalline yttrium aluminate garnet, magnesium aluminate spinel and α-alumina, Journal of Nuclear Materials 274 (1999) 78-83.

Captions

Table 1 Irradiation condition (HFR-Petten) and bending strength results on SiC/SiC and SAY joined SiC/SiC

(* irregular failure, not in compliance with the standard ASTM C1341)

Table 2 The average chemical composition of SAY before and after irradiation at LVR-15 measured by EDS

Table 3 The average chemical composition of SAMg before and after irradiation at LVR-15 measured by EDS

Table 4 Irradiation effects on glasses and glass-ceramics.

Figure 1 Visual appearance of SAY pellets before (a) and after (b) irradiation at LVR-15.

Figure 2 SEM microscopy of SAY before (a) and after (b) irradiation at LVR-15: SEM-EDS on irradiated and non-irradiated SAY pellets confirms the presence of three phases: white phase ($Y_2Si_2O_7$), grey phase (Al_2SiO_5) and black phase (SiO_2)

Figure 3 Visual appearance of SAMg pellets before (a) and after (b) irradiation at LVR-15.

Figure 4 SEM microscopy of SAMg before (a) and after (b) irradiation at LVR-15

Figure 5 SEM of SAY joined SiC/SiC (type 3) before (a) and after (b) irradiation and bending test (HFR-Petten) at 600 °C, $16.3 \cdot 10^{24}$ n/m^2

Figure 6 Microstructure by SEM of SAY in SAY joined SiC/SiC before (a) and after (b) irradiation (HFR-Petten) at 600 °C, $16.3 \cdot 10^{24}$ n/m^2

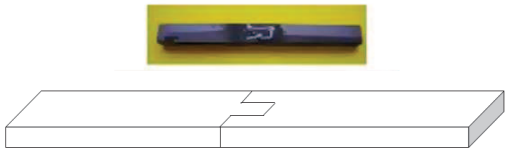
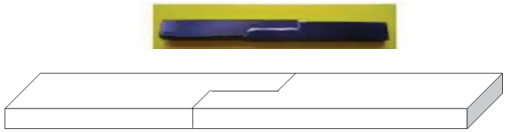
	Before irradiation	After irradiation (HFR-Petten, NL)		
	Bending strength [MPa]	550 ℃ 9-11 10^{24} n/m^2	600 ℃ 16-22 10^{24} n/m^2	820 ℃ 31-32 10^{24} n/m^2
As received, not joined SiC/SiC	418 ± 45	304	*113 ± 14 (*)*	*59 ± 17 (*)*
Not joined SiC/SiC (1420 ℃, 30 min, Ar, 1240℃ 1h)	283 ± 8	-	-	-
SAY Joined SiC/SiC type2 	122 ± 10	-	118	89±17
SAY Joined SiC/SiC type3 	149	-	65	-

Table 1 Irradiation condition (HFR-Petten) and bending strength results on SiC/SiC and SAY joined SiC/SiC

(* irregular failure, not in compliance with the standard ASTM C1341)

Element	O	Al	Si	Y
Non-irradiated sample SAY	46.52	8.13	26.13	19.22
Irradiated sample SAY	46.83	8.23	24.98	19.96

Table 2 The average chemical composition of SAY before and after irradiation at LVR-15 measured by EDS

Element	O	Mg	Al	Si
Non-irradiated sample SAMg	51.88	5.16	14.78	28.18
Irradiated sample SAMg	51.01	5.23	15.06	28.70

Table 3 The average chemical composition of SAMg before and after irradiation at LVR-15 measured by EDS

Glass or glass-ceramic	Irradiation conditions	Effects	Reference
Crystalline silica	$2x10^{24}$ n/m^2	15 vol% swelling	[12, 13]
Vitreous silica	$2x10^{24}$ n/m^2	3 vol% shrinkage	[12, 13]
Glass-ceramic (MACOR, Corning) 50 % vol. fluorophlogopite mica crystals ($KMg_3AlSi_30_{10}F_2$) embedded in a borosilicate glass matrix	room temperature, $1x10^{20}$ - $1x10^{22}$ n/m^2 (°)	No significant. These studies show that there exists no permanent degradation in measured mechanical and electrical properties of MACOR at doses as large as 1Q22 14 MeV n/m^2.	[18]
Eight glass-ceramics containing $KMg_3AlSi_30_{10}F_2$ or $Li_2Si_2O_5$ in a glass matrix	400-550 °C, $2.4x10^{22}$ n/m^2 (°)	General resistance to changes in thermal expansion and most did not experience severe loss of mechanical integrity. The maximum volume expansion occurred in several of the fluorophlogopite-based glass ceramics (3.0%).	[19]
MACOR glass-ceramic	room temperature, $4x10^{22}$ -$1x10^{23}$ n/m^2 (°)	expansion of the mica phase and contraction of the glass matrix	[20]
SAY, SAMg glass-ceramics	50 °C, 6.92×10^{24} n/m^2 (*)	darkening	This work
SAY, SAMg glass-ceramics (used to join SiC/SiC)	550 °C 9-11 .10 ^{24} n/m^2 600 °C, 16-22x10 ^{24} n/m^2 (**)	none	This work

Table 4 Irradiation effects on glasses and glass-ceramics.

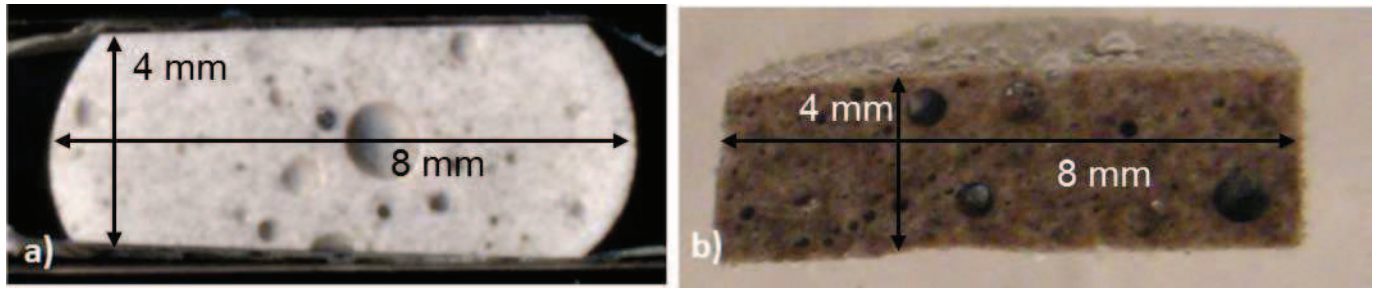
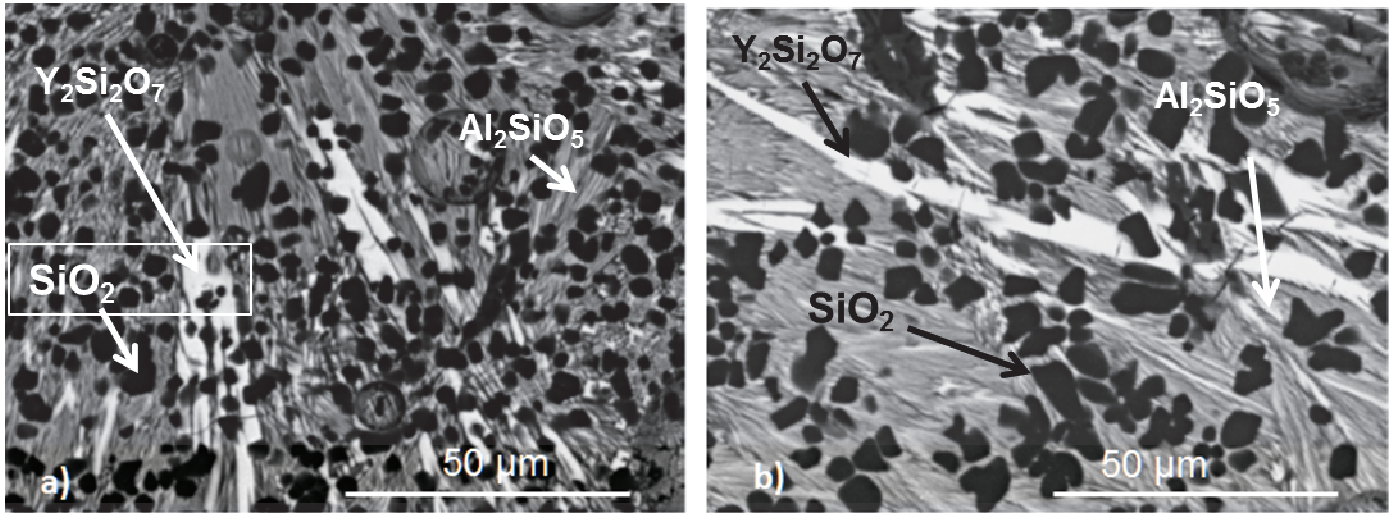


Figure 1 Visual appearance of SAY pellets before (a) and after (b) irradiation at LVR-15.



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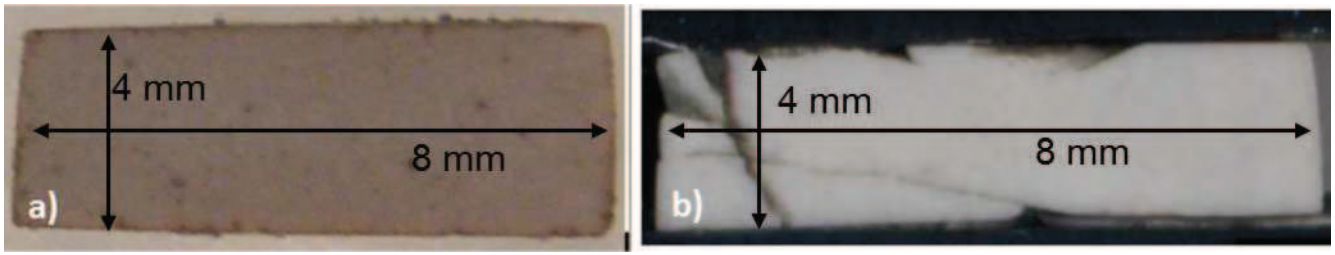

Figure 3 Visual appearance of SAMg pellets before (a) and after (b) irradiation at LVR-15.

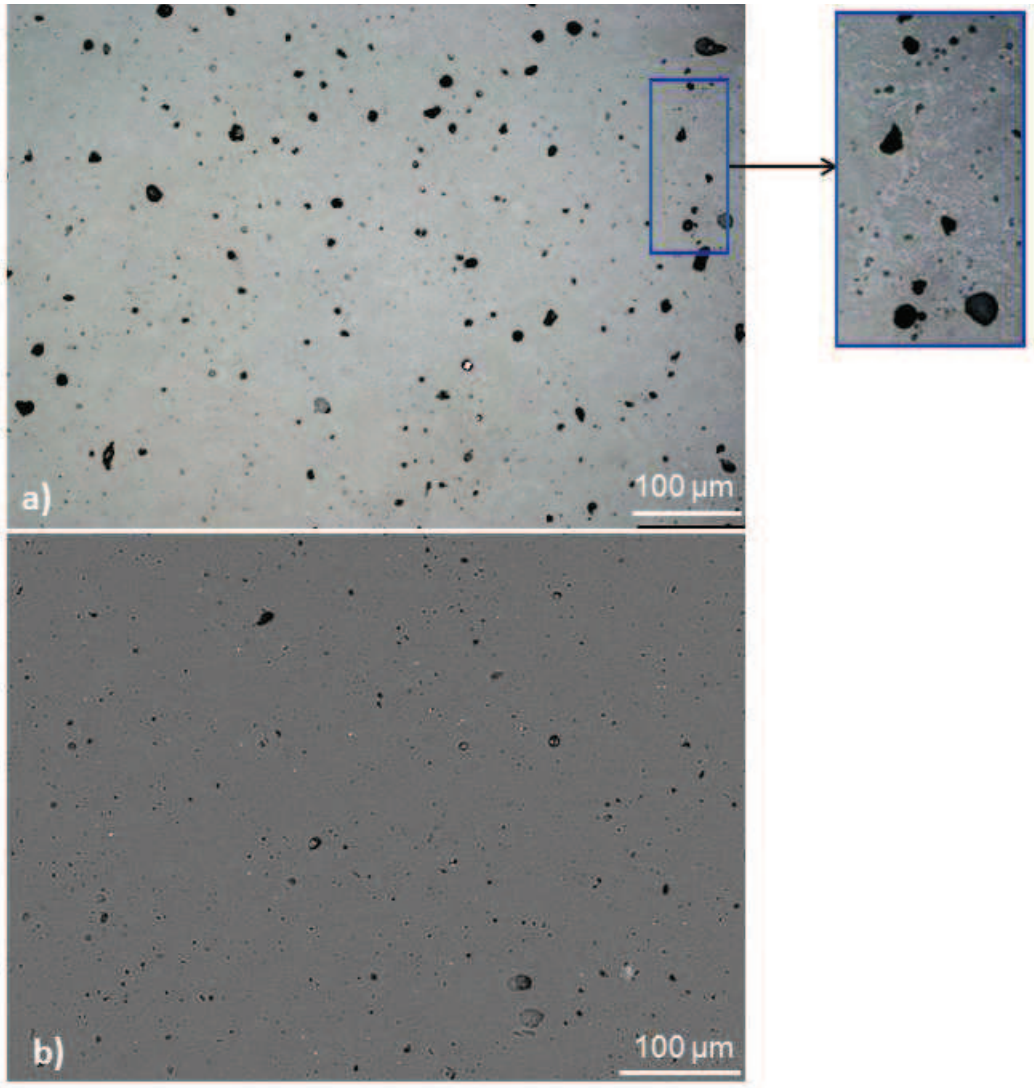

Figure 4 SEM microscopy of SAMg before (a) and after (b) irradiation at LVR-15

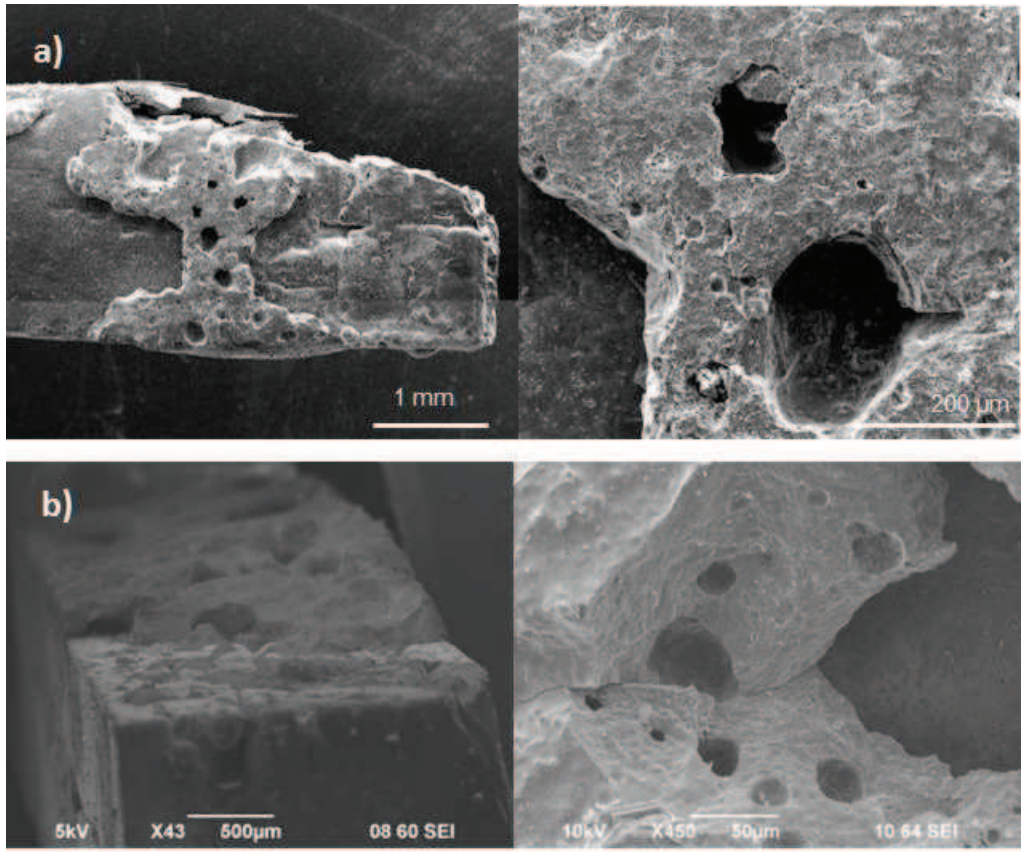

Figure 5 SEM of SAY joined SiC/SiC (type 3) before (a) and after (b) irradiation and bending test (HFR-Petten) at 600 °C, $16.3 \cdot 10^{24}$ n/m^2

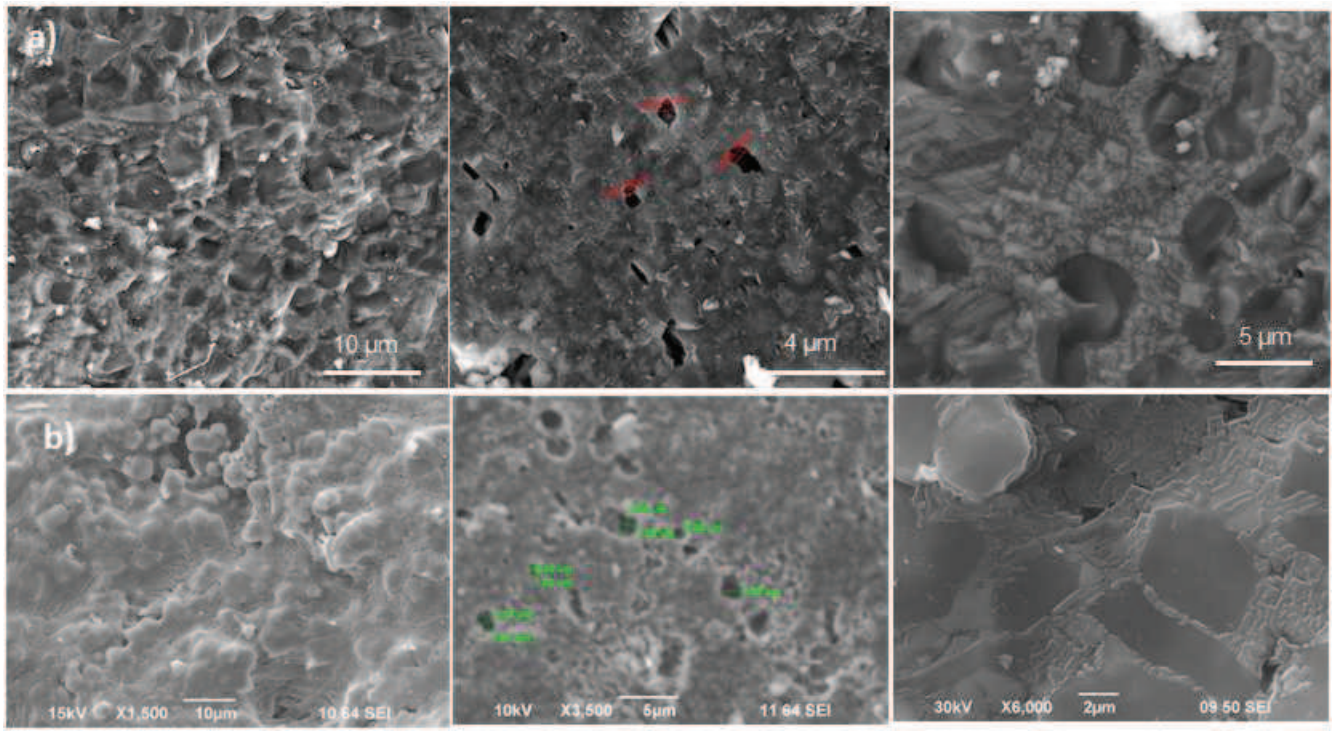

Figure 6 Microstructure by SEM of SAY in SAY joined SiC/SiC before (a) and after (b) irradiation (HFR-Petten) at 600 °C, $16.3 \cdot 10^{24}$ n/m^2